

# KONDO /HEAVY FERMION SYSTEMS

## dHvA Study on CeNiSn

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In recent years, attention has been focussed on the Kondo insulators or Kondo semimetals such as CeNiSn, CeRhSb,  $\text{Ce}_3\text{Bi}_4\text{Pt}_3$ ,  $\text{YbB}_{12}$ , and  $\text{SmB}_6$ . Among them, CeNiSn crystallizes with an orthorhombic structure. The small pseudogap formation was clearly observed in the specific heat and NMR measurements. Moreover, many anisotropic physical properties were reported in CeNiSn, reflecting the crystal structure. The result of magnetoresistance suggested that the anisotropic pseudogap is easily closed by applying a large field along the a-axis. Therefore, it is important to obtain clear evidence for the existence of the Fermi surface by the dHvA effect, under these conditions.

We have performed the dHvA experiments on CeNiSn in pulse field up to 50 T and at temperature down to 0.4 K for the principal field directions, namely along a, b, and c-axis. This is very challenging since the dHvA experiment requires high quality single crystals; although it is extremely difficult to grow the crystals of CeNiSn.

Unfortunately, no reliable data was obtained. This is mainly due to the poor quality of the sample, which suppresses the dHvA oscillation.

## High Field c-axis Magnetotransport of Single Crystal $\text{YbNi}_2\text{B}_2\text{C}$

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The intermetallic borocarbides,  $\text{RENi}_2\text{B}_2\text{C}$  (RE=Dy-Lu, Y), have generated great interest due to their display of such diverse phenomena as superconductivity, magnetic order, and correlated electron behavior.<sup>1</sup>  $\text{YbNi}_2\text{B}_2\text{C}$  is the only compound in the series that does not order or become superconducting down to 50 mK.<sup>2</sup>  $\text{YbNi}_2\text{B}_2\text{C}$  exhibits strongly correlated electron behavior at low temperature with a Sommerfeld coefficient of 530 mJ/molK<sup>2</sup>.<sup>2</sup> Specific heat measurements indicate an estimated Kondo temperature ( $T_K$ ) of ~10 K, while inelastic neutron measurements indicate a  $T_K$  of ~3.8 K.<sup>2,3</sup> Strong anisotropy was observed in previous investigations of the magnetotransport of  $\text{YbNi}_2\text{B}_2\text{C}$ , when the magnetic field (H) was applied parallel or perpendicular to the c-axis.<sup>4</sup> However, in none of these studies was the current applied along the c-axis.

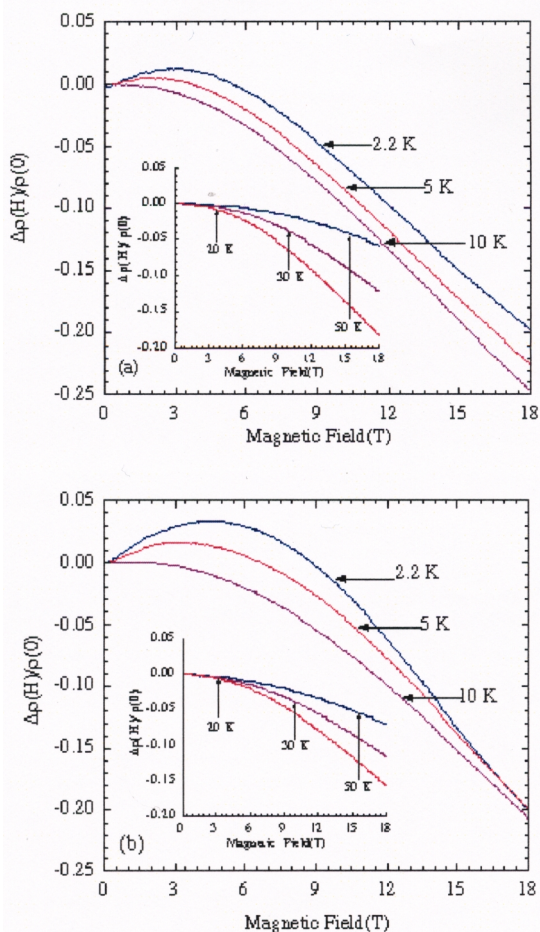
We have measured c-axis magnetotransport properties of the tetragonal  $\text{YbNi}_2\text{B}_2\text{C}$  compound down to 2 K and up to 18 T at the NHMFL/LANL. Transverse and longitudinal magnetoresistance have been measured with current applied along the c-axis of the tetragonal structure. A well-defined maximum in the magnetoresistance is observed at temperatures below 10 K at approximately 5 T. At higher temperatures, the magnetoresistance is always negative and weakens as the temperature is increased.

<sup>1</sup> Canfield, P.C., *et al.*, Physics Today, **51**, 40 (1998).

<sup>2</sup> Lacerda, A.H., *et al.*, Philos. Mag. B, **74**, 641 (1996); Yatskar, A., *et al.*, Phys. Rev. B **54**, R3772 (1996).

<sup>3</sup> Sierks, C., *et al.*, Physica B, **259-261**, 592 (1999).

<sup>4</sup> Yatskar, A., *et al.*, Phys. Rev. B, **60**, 8012, and references there in.



**Figure 1.** The normalized magnetoresistance,  $\Delta\rho(H)/\rho(0)=[\rho(H,T)-\rho(0,T)]/\rho(0,T)$ , as a function of the applied magnetic field for  $\text{YbNi}_2\text{B}_2\text{C}$ . In both (a) and (b), the current is applied along the c-axis. In (a), the magnetic field is applied parallel to the c-axis, and in (b) the magnetic field is applied perpendicular to the c-axis. The insets in (a) and (b) show the high temperature normalized magnetoresistance.

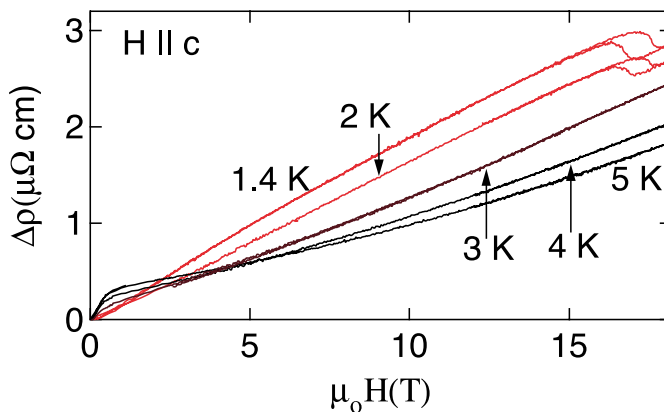
## High Field Magnetotransport of a New Class of Ce-Based, Heavy Fermion Superconductors: $\text{CeRhIn}_5$

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$\text{CeRhIn}_5$  has recently attracted attention due to unusual pressure-induced superconductivity.<sup>1</sup> For example, in  $\text{CeIn}_3$  when pressure is applied the antiferromagnetic transition moves smoothly toward zero until the onset of superconductivity appears.<sup>2</sup>

In  $\text{CeRhIn}_5$ , an antiferromagnetic transition occurs at ambient pressure at 3.8 K. Upon application of pressure, the resistive signature of the antiferromagnetic transition moves upward in temperature and becomes weaker, until disappearing completely above 14.5 kbar. Increasing the pressure to 16.3 kbar results in superconductivity at 2.1 K.<sup>1</sup>  $\text{CeRhIn}_5$  may be considered a quasi-2d structure with alternating layers of  $\text{CeIn}_3$  and  $\text{RhIn}_2$ . Specific heat measurements indicate an enhanced Sommerfeld coefficient of  $\sim 400 \text{ mJ/mol K}^2$  below 10 K.<sup>1</sup> Neutron scattering measurements indicate the magnetic moments lie in the basal plane of the tetragonal structure with a spiral along the c-axis with a pitch of 0.297.<sup>3</sup>

In an attempt to better understand the behavior of  $\text{CeRhIn}_5$ , we have made use of the 20 T superconducting magnet at the Pulse Field Facility to measure the resistivity as a function of temperature and magnetic field. At low temperature, we find a positive linear behavior in the magnetoresistance. At 1.4 K, the magnetoresistance displays hysteresis centered at  $\sim 17.5 \text{ K}$  when the magnetic field is applied along the c-axis. At higher temperatures, we find a positive peak in the magnetoresistance that becomes weaker until the magnetoresistance displays only a negative contribution at high temperatures.



**Figure 1.**  $\Delta\rho$  ( $\rho(H,T)-\rho(0,T)$ ) versus magnetic field at low temperatures.  $\Delta\rho$  is nearly linear at the temperatures displayed here. At the lowest temperatures, hysteresis is observed. The upper side of the hysteresis loop corresponds to an increasing magnetic field.

<sup>1</sup> Hegger, H., *et al.*, Phys. Rev. Lett., **84**, 4986 (2000).

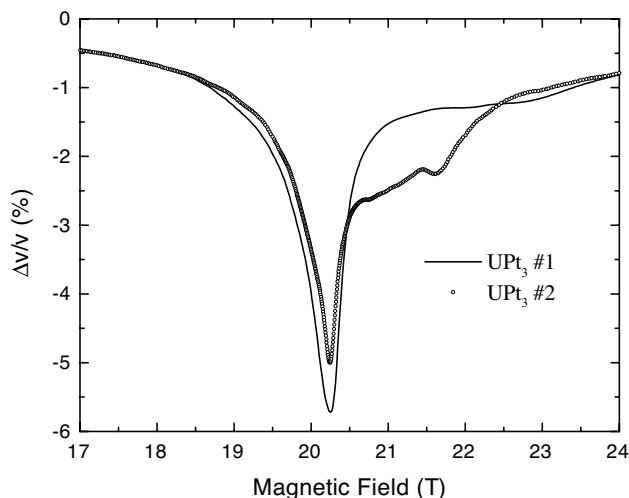
<sup>2</sup> Walker, I.R., *et al.*, Physica C, **282-287**, 303 (1997) and references therein.

<sup>3</sup> Bao, Wei, *et al.*, to be published.

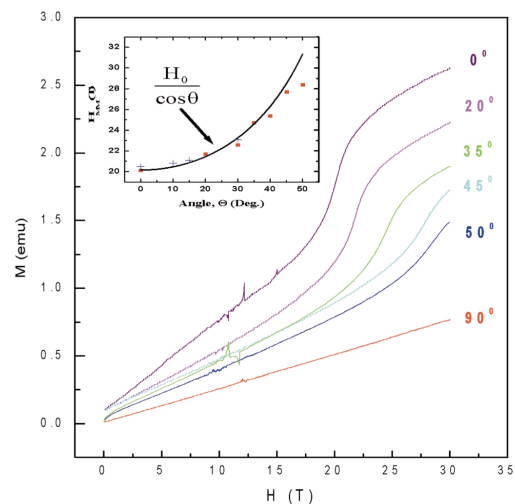
## Ultrasonic and Magnetization Measurements in $\text{UPt}_3$ Near the Metamagnetic Transition

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High-resolution velocity and magnetization measurements have been carried out on samples of  $\text{UPt}_3$  in fields up to 33 T and temperatures down to 40 mK. Magnetization measurements were done on a Lake Shore vibrating sample magnetometer (VSM), on a spherical sample, and a rectangular sample (sample #1). Velocity measurements were done on two samples (sample #1 and #2; sample #2 is believed to be of a higher quality, as evidenced from a higher  $T_c$ ). We had reported earlier,<sup>1</sup> data on measurements in sample 2, where we saw a second velocity dip at a higher field, and quantum oscillations near the transition at lower temperatures. Fig 1. shows the velocity data on the two different samples. The second velocity peak is not that pronounced, and also quantum oscillations were not seen, suggesting that these effects could be more enhanced in the purer sample.



**Figure 1.** Comparison of the velocity shift at ~70 mK in samples #1 and #2.



**Figure 2.** Magnetization of  $\text{UPt}_3$  for various orientation of the magnetic field. The inset shows anisotropy of the metamagnetic feature. The angles listed are measured from the basal plane.

VSM measurements were done on the spherical sample and sample 1. Fig. 2. shows the increase in magnetization at the metamagnetic transition. The measurements were done for various orientations of the magnetic field with respect to crystal axis. The inset shows the anisotropy of this effect. The solid curve is a fit proportional to  $(\cos\theta)^{-1}$ .

**Acknowledgements:** This work was supported by NSF DMR-9704020 and DMR-9971123.

<sup>1</sup> Feller, J.R., *et al.*, Phys. Rev. Lett., **B62**, 11538-11544 (2000).

## First-Order Valence Transition in $\text{YbInCu}_4$ in the (B,T)-Plane

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For years, the most famous example of the first order transition into a state with an intermediate valence has been the isostructural  $\gamma - \alpha$  transition in metallic Ce. (Changes in the valence state are usually judged by the change in the unit cell volume or by spectroscopic means, which strictly speaking do not always provide exact valence values). Although a structural transition in a crystalline matter possessing

a critical point in the (P,T)-plane is of a great interest by itself, an observation of the  $\gamma$  -  $\alpha$  transition in Ce has, in addition, created a whole new field of studying intermediate or mixed valence states in rare earths and actinides (both for elemental metals and intermetallic compounds).

While  $\gamma$  -  $\alpha$  transition in Ce takes place in the pressure range P in 10 to 20 kbar, the isostructural transition in YbInCu<sub>4</sub> is observed at  $T_v \approx 40$  K and at ambient pressure (the major experimental results are best summarized in Ref. 1). Looking akin to the Ce “isomorphic” transition, it has attracted a lot of interest recently due to the possibility of study of that phenomenon in much greater details.

We addressed the issue of the phase diagram of YbInCu<sub>4</sub> in the (B,T)-plane, where B is a magnetic field, T is a temperature. Indeed, among many interesting results (see reference 1), the most surprising one is the universality of the first order transition line for YbInCu<sub>4</sub> and its alloys. Namely, being expressed in the reduced variables ( $B/B_{\{c0\}}$ ,  $T/T_{\{v0\}}$ ), the transition line separating the high temperature phase (paramagnetic, local moments) and the low temperature “metallic” phase, is a perfect circle (here  $T_{\{v0\}}$  is the structural transition temperature in the absence of the magnetic field and  $B_{\{c0\}}$  is the critical field at T=0).

We have shown that these results are surprisingly well described in terms of an entropy first order phase transition between the local f-moments phase and another phase probably of a less ordinary nature.<sup>2</sup> An origin of this phase however, seems not to be important if this second phase is characterized by a larger energy scale. If the same ideas were applied to the  $\gamma$  -  $\alpha$  transition in Ce, it would have predicted a similar phase diagram in high magnetic fields with  $B_{c0} \approx 200$  T. This is the field range achievable for modern megagauss magnetic field experiments. A characterization of the  $\gamma$  -  $\alpha$  transition in Ce not only in the (P,T)-plane, but also in the (B,T)-plane thus looks feasible and presents a great interest.

<sup>1</sup> Sarrao, J.L., Physica B, **259**, 128 (1999).

<sup>2</sup> Dzero, M.O., *et al.*, J. Phys.: Condens. Matter, **12**, L1 (2000).

## Correlated Electron Materials

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Our research in correlated electron materials has investigated further heavy Fermion superconductors, hexaborides, and Kondo insulators.

(1) We have been involved in two areas of heavy Fermion superconductivity. In collaboration with the group of H.R. Ott, the specific heat at low temperatures of UBe<sub>13</sub> in applied magnetic fields has been studied. A particular form of scaling of the data in applied fields further strengthens the argument for unconventional superconductivity in this material. Extensive studies in collaboration with J.D. Thompson and J.L. Sarrao at LANL on the CeTIn<sub>5</sub> (T=Co, Rh or Ir) materials establishes a new family of heavy Fermion superconductors, including the only new such family of Ce-based materials superconducting at ambient pressure since the discovery of the original heavy Fermion superconductor, CeCu<sub>2</sub>Si<sub>2</sub>, by Steglich in 1979. Successful growth of single crystals of the Co material by Petrovic set a new maximum for  $T_c$  for heavy Fermion superconductivity of 2.3 K. The angular dependence of the upper critical field of CeCoIn<sub>5</sub> has characteristics similar to quasi-2D materials. The interesting question being pursued in these experiments is the extent to which the materials are quasi-2D. Goodrich and Hall have investigated the Fermi surface of these materials in deHaas-van Alphen experiments, and it has proved possible as well to study the evolution of Fermi surface properties across the whole alloy system with La substitution for Ce in the Rh system. Similar studies are in progress for the other systems.

(2) Extensive work on hexaborides has progressed in collaboration with the group of H.R. Ott at ETH Zurich. There have been three thrusts



to this experimental work. In the first, we have investigated optically (measurements of Degiorgi), and through transport measurements, the extent to which the doped excitonic insulator model for the weak ferromagnetism describes the data. We have so far no strong confirmation of this model, and the transport data supports an impurity band picture, consistent with new GW band structure results finding a large rather than small direct gap in  $\text{SrB}_6$ . Secondly, studies have been continued on  $\text{EuB}_6$  and its alloys. A very large Kerr rotation has been found at low temperature in this material, adding to the unusual low temperature properties studied earlier. And thirdly, a successful protocol for growth of single crystals of  $\text{YbB}_6$  has been developed by Bianchi, allowing now for comparative studies of this divalent hexaboride with the other known divalent hexaborides. Preliminary results find weak ferromagnetism in this material.

- (3) Kondo insulators, doped-Kondo insulators and related half Heusler compounds have been investigated. A collaboration with DiTusa and Aeppli on doped FeSi has discovered the presence of magnetoresistance arising from quantum interference. Studies have also been made to determine the nature of small moment ferromagnetism in single crystals of pure FeSi. Single crystal studies of  $\text{TiCoSb}$  have been made. This appears to be a metallic compound which becomes semiconducting with Fe-alloying at the few percent level. The doped Fe in this material carries a moment of approximately  $0.6 \mu_B$ , obeying a Curie law out to the limit of Fe-substitution that we can grow crystals of,  $\text{TiCo}_{0.2}\text{Fe}_{0.8}\text{Sb}$ . The non-interacting nature of the moments found is reminiscent of what has been found in the Kondo insulators and our studies are directed at establishing this correspondence in the half Heusler materials.

## Electronic Structure of $\text{RMIn}_5$ ( $\text{R}=\text{Ce, La, Y}$ ; $\text{M}=\text{Co, Ir, Rh}$ )

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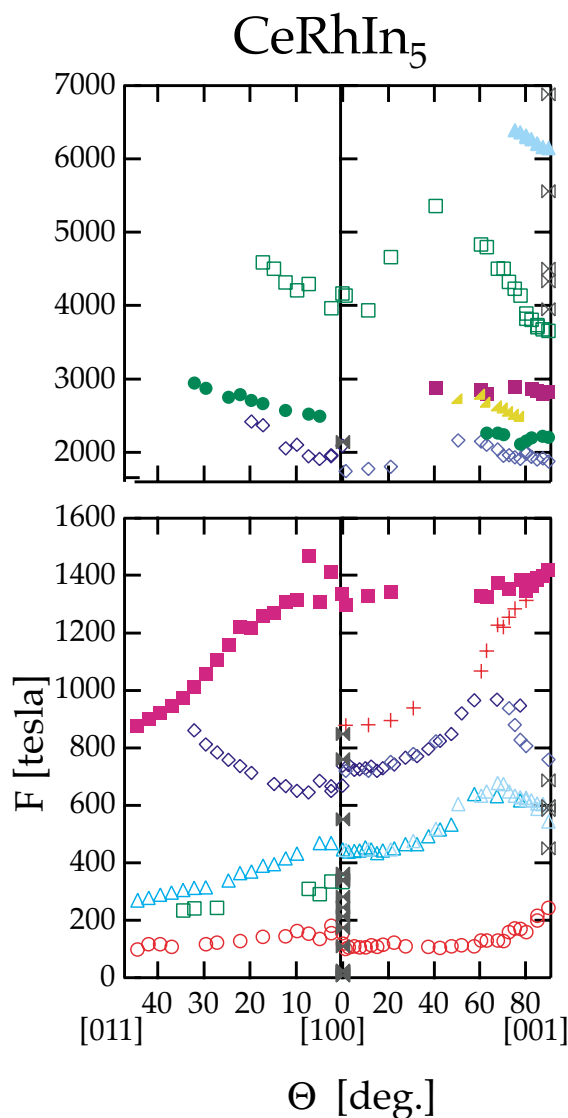
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During the past year, we have conducted measurements on a family of materials with a common crystallographic structure: the quasi-2D



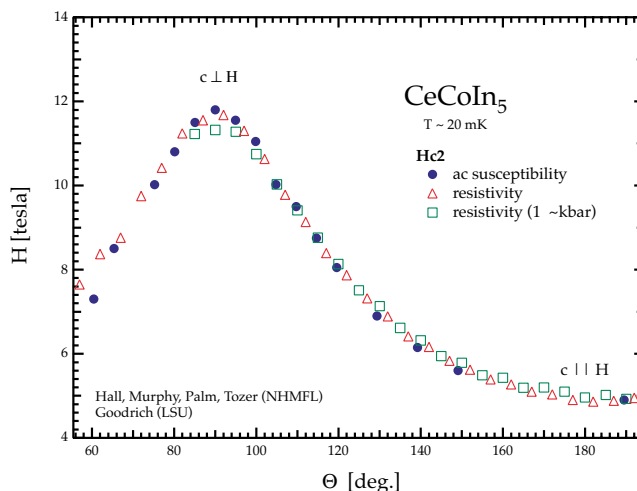
**Figure 1.** The measured frequencies in  $\text{CeRhIn}_5$  plotted against angle.

$\text{H}_0\text{C}_0\text{Ga}_5$  structure (space group  $\text{P4}/\text{mmm}$ ). Interest in the heavy-fermion and magnetic Ce doped variants began when  $\text{CeRhIn}_5$  was found to be superconducting under pressure ( $P_c=16$  kbar) with  $T_c=2.1$  K.  $\text{CeIrIn}_5$  proved to be an ambient pressure superconductor with a  $T_c$  of 0.4 K.  $\text{CeCoIn}_5$  has the highest  $T_c$  at ambient pressures, 2.8 K. The purpose of our studies has been to fully characterize the electronic structure of these materials in an effort to understand the interplay of magnetism and superconductivity.

A complete fermiological study was conducted on  $\text{CeRhIn}_5$ , the results of which are reported in a forthcoming article. We confirmed the quasi-2D nature of this material using the de Haas van Alphen (dHvA) effect to map the Fermi surface (FS) as a function of angle with respect to applied magnetic field. The range of masses for the measured orbits is  $3m_e$  to  $6m_e$ . Based on a comparison of our results with a calculation of the band structure, we found (1) the FS consists of structures arising from two hole bands and one electron band, and (2) there are spin density waves along the c-axis that give rise to the anti-ferromagnetism in the a-b plane and distort the FS from that predicted by theory. Fig. 1 shows a rotation plot of the measured dHvA frequencies. In addition, dHvA pressure measurements have been initiated to elucidate changes in the FS as the superconducting phase is approached from the antiferromagnetic ground state.

A considerable amount of effort has been put into the measurement of  $\text{CeCoIn}_5$ . The results of this study will be the subject of at least two separate papers, one detailing the 2D superconducting behavior as a function of temperature and angle (for example see Fig. 2) along with measurements of a possible new low temperature magnetic phase in the mixed state. The second paper will provide detailed measurements of the FS.

Another project has been to quantify the evolution of the FS as a function of chemical doping. Ce atoms in  $\text{CeRhIn}_5$  were replaced with La. An entire series of dHvA measurements have been carried out on these Ce-LaRh115 alloys. This work is the subject of a Ph. D. thesis to be written by U. Alver, a graduate student at Louisiana State University.



**Figure 2.** The critical field,  $H_{c2}$ , in  $\text{CeCoIn}_5$  plotted against angle at 25 mK.

dHvA measurements on the normal metals  $\text{LaRhIn}_5$ ,  $\text{LaIrIn}_5$ , and  $\text{YCoIn}_5$  provide the fermiologist an opportunity to examine FS orbits near the quantum limit.  $\text{LaRhIn}_5$  has a small piece of FS ( $f \sim 7$  T) that persists up to 60 K at fields up to 30 T. Above the  $n=1$  Landau level (after the 7 T pocket is depopulated), we observe that  $\text{LaRhIn}_5$  becomes weakly magnetic, which may indicate FS polarization. Additionally, rotational studies reveal a FS geometrically similar to their heavy fermion cousins, exhibiting evidence of a quasi-2D structure.

## Locally Critical Quantum Phase Transitions in Heavy Fermion Systems

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The conventional theory of quantum phase transitions assumes that the only important low-energy degrees of freedom are long-wavelength fluctuations of the order parameter. However, this picture has been contradicted by recent experiments on heavy fermion metals at a zero-temperature phase transition. In particular, neutron scattering from  $\text{CeCu}_{6-x}\text{Au}_x$  has revealed an anomalous frequency and temperature dependence over essentially the entire Brillouin zone, suggesting that the dynamics at atomic length scales are also critical.<sup>1</sup>

We show that a dynamical spin susceptibility, just as observed experimentally, can arise at a new type of *locally critical* quantum phase transition, where local critical degrees of freedom coexist with spatially extended ones.<sup>2</sup> We reach this conclusion by solving the Kondo lattice model within an extended dynamical mean-field formalism.<sup>3</sup> The model exhibits a zero-temperature transition between a paramagnetic metal and an antiferromagnetic metal. The nature of this transition depends on the effective dimensionality of spin fluctuations in the model. Three-dimensional fluctuations produce a conventional transition, whereas quasi-two-dimensional fluctuations (such as those occurring in  $\text{CeCu}_{6-x}\text{Au}_x$ ) give rise to locally critical physics. We believe that the phenomenon of local criticality is of general relevance to strongly correlated systems. It might explain, for example, some of the puzzling features of the high-temperature superconductors.

<sup>1</sup> Schröder, A., *et al.*, Phys. Rev. Lett., **80**, 5623-5626 (1998).

<sup>2</sup> Si, Q., *et al.*, cond-mat/0011477.

<sup>3</sup> Si, Q., *et al.*, Phys. Rev. B, **61**, 5184-5193 (2000).

## de Haas van Alphen Experiments in Ce and Yb Compounds

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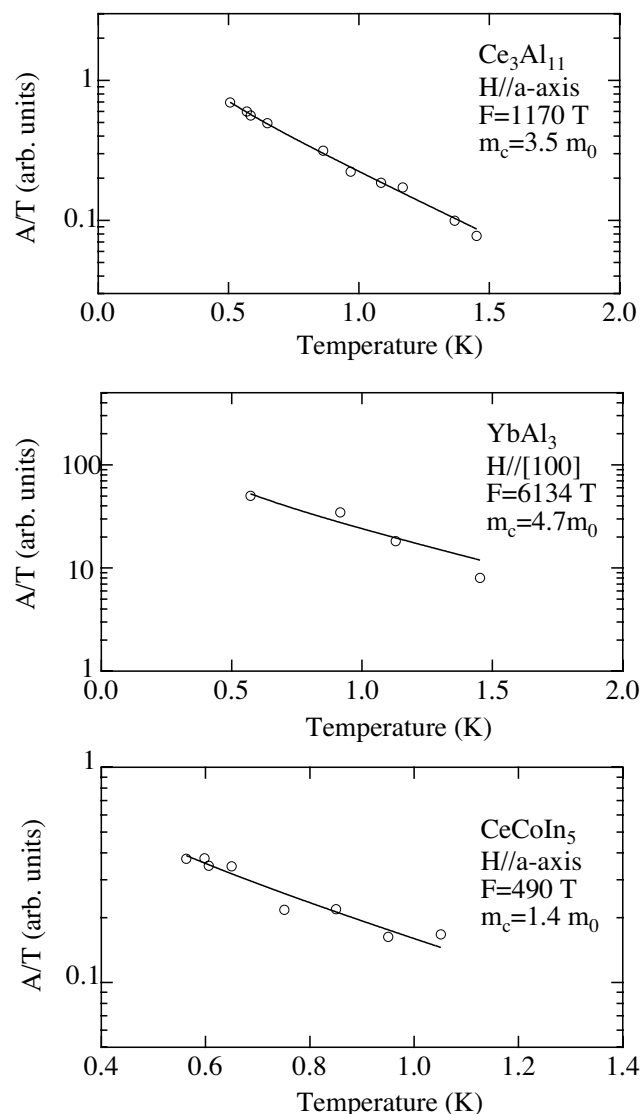
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We measured the de Haas van Alphen (dHvA) signal of several Ce and Yb compounds using the 60 T short pulse magnet at the LANL/NHMFL facility:

1. The orthorhombic compound  $\text{Ce}_3\text{Al}_{11}$  exhibits incommensurate antiferromagnetism below  $T_N = 3.2$  K; the critical field for the metamagnetic transition to a ferromagnetically aligned state is 2 T.<sup>1</sup> We measured the high field dHvA signal as part of a larger effort to determine how the Fermi surface



**Figure 1.**  $A/T$  vs. temperature  $T$ , where  $A$  is the amplitude of the dHvA signal, for  $\text{Ce}_3\text{Al}_{11}$ ,  $\text{YbAl}_3$  and  $\text{CeCoIn}_5$ . The frequency of the signal, and the effective mass deduced from the slope of these plots, are given in the figure.

changes due to the metamagnetic transition. For field along the a-axis, we detected a signal at a frequency of 1170 T. The temperature dependence of the amplitude (Fig. 1) implies an effective mass  $m^* = 3.5m_0$  for this branch. This is the first report of a dHvA signal in this compound; we will follow up this work with extensive studies of the Fermi surface in low, DC fields.

2. The compound  $\text{YbAl}_3$  exhibits mixed valence, and a large Kondo temperature. The Fermi surface has been measured in low fields, using DC dHvA techniques.<sup>2</sup> In an effort to determine how the Fermi

surface changes with magnetic field, we performed the high field dHvA measurement for field parallel to [100]. We detected a signal at a frequency of 6134 T, which corresponds to the frequency 6510 T seen in the low field measurement. The effective mass determined at high field is  $4.7m_0$ , considerably smaller than the value  $19m_0$  reported at low field. The high field signal was very noisy, and the signal amplitude depended strongly on the field interval chosen for the fast Fourier transform, so further work will be necessary to establish whether this apparent field dependence is real.

3. The tetragonal compound  $\text{CeCoIn}_5$  is one of a newly discovered class of heavy fermion superconductors. We measured dHvA on a sample grown in In flux. For field along the a-axis, we observed a dHvA signal at 490 T with a corresponding effective mass of  $1.4m_0$ . This frequency was not observed in recent dHvA experiments using low ( $<20$  T) DC fields.<sup>3</sup> The possibility that the signal arises from free indium seems to be ruled out by the fact that the effective masses observed in  $\text{In}^4$  are all five times smaller than the value observed here. A possible way to confirm this result would be to use low fields and a field modulation technique.

<sup>1</sup> Muñoz, A., et al., J. Phys.: Condens. Matter, 7, 8821-8831 (1995).

<sup>2</sup> Ebihara, T., et al., Physica B, 281&282, 754-755 (2000).

<sup>3</sup> Donovan Hall, private communication.

<sup>4</sup> Hughes, A.J., et al., J. Phys. C (Solid State Phys.), 2, 661-672 (1969).

## Griffiths Phase of the Kondo Insulator Fixed Point

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Heavy fermion compounds have long been identified as systems which are extremely sensitive to the presence of impurities and other imperfections. In recent years, both experimental and theoretical work has demonstrated that such disorder can lead to unusual, non-Fermi liquid behavior for most physical quantities.

Based on an initial success in explaining the anomalous behavior of  $\text{UCu}_{5-x}\text{Pd}_x$  by means of the so-called Kondo disorder model,<sup>1</sup> we have recently extended the analysis to describe Anderson localization effects in a strongly correlated environment.<sup>2</sup> Using this approach, we have demonstrated<sup>2</sup> that the emergence of NFL properties can be described in terms of a quantum Griffiths phase induced by Anderson localization effects. Furthermore, the onset of anomalous behavior occurs already at relatively weak disorder, an effect we ascribe to the *proximity to the Kondo insulator fixed point*. In a clean compound, unitary Kondo scatterers act coherently to create a hybridization gap and the state is the familiar Kondo insulator. A small deviation from unitarity leads to the formation of heavy fermionic quasi-particles. In a disordered system, however, large spatial fluctuations induce the appearance of random unitary scatterers, or “Kondo insulator droplets” which are responsible for a strong *renormalized effective disorder*. These droplets, in turn, are regions of depleted density of states (DOS) which fail to quench nearby localized moments. The latter ultimately lead to the NFL behavior.

Our results demonstrate that the localization-based scenario provides a very robust and quantitatively relevant mechanism for the NFL behavior. Since the relevant fluctuations must be present in any moderately disordered system, this route should be of direct relevance to most disordered heavy fermion compounds, irrespective of the proximity to any magnetic ordering. Our key point is that the corresponding Griffiths phase is a direct consequence of the proximity not to any magnetic, but rather to the Kondo insulator fixed point. The deviation from the Kondo insulator provides an energy scale which is universally small for any heavy fermion metal, since it is defined by the underlying Kondo energy. This observation also provides a simple explanation for the notorious sensitivity of HF systems to disorder, a feature which we believe is at the origin of all the observed anomalies.

<sup>1</sup> Miranda, E., et al., Phys. Rev. Lett., **78**, 290 (1997).

<sup>2</sup> Miranda, E., et al., Phys. Rev. Lett., **86**, 264(2001); to appear in J. Magn. Magn. Matter (2001).



## Heat Capacity Measurements in NHMFL 60 T Quasi-Continuous Magnet

■ IHRP ■

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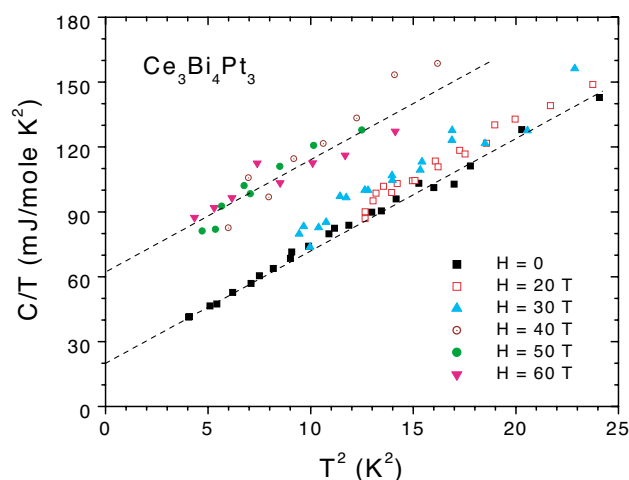
Stewart, G., UF, Physics

We have successfully demonstrated the feasibility of the specific heat measurements in pulsed magnetic fields up to 60 T in the NHMFL/LANL 60 T Long Pulse (60 T LP) Magnet, the primary objective of our project.

We used a quasiadiabatic heat pulse method that is based on delivering a known amount of heat to the sample, and measured its temperature before and after the heat pulse. It requires that the complete assembly of the sample, heater, and thermometer comes to thermal equilibrium before and after the heat pulse. This condition was impossible to satisfy in the short pulse (capacitor bank driven) magnets, where the *total* magnetic field pulse was on the order of ten milliseconds. The development of the 60 T long-pulse magnet at LANL, with the 100 millisecond *flat top*, opened the door for the equilibrium measurements in pulsed magnetic fields.

Working in a pulsed field environment necessitated development of a specialized probe. Made of plastic materials to avoid eddy current heating, it allows us to perform heat capacity measurements at temperatures between 1.6 K and 20 K (at present we are battling the systematic error in temperature above 4 K, increasing up to 20 % at 20 K) in fields up to 60 T. To maximize the available experimental space, a novel vacuum tapered seal was developed. The conical plug part of the joint is made out of G-10, and the matching vacuum can is made out of 1266 Stycast epoxy. The differential thermal contraction between the parts aids in producing a superfluid-tight joint. The simple construction of the joint resulted in a comparatively large (16 mm diameter) experimental region.

The quasiadiabatic method turns out to be applicable to a rather large variety of metallic and insulated compounds. The primary scientific result from that effort was observation of the insulator-to-metal transition in the Kondo Insulator,  $\text{Ce}_3\text{Bi}_4\text{Pt}_3$ , published in *Nature*, vol. **405**, 160 (2000), where we saw a threefold increase in the zero temperature Sommerfeld coefficient  $\gamma_H = C_H/T|_{T \rightarrow 0}$ , between the fields of 30 and 40 T. Other systems that were shown to be amenable to investigation with the heat-pulse method include valence-transition compound,  $\text{YbInCu}_4$ , heavy fermions (such as  $\text{UCd}_{11}$ ,  $\text{UBe}_{13}$ ), and insulating compounds.



**Figure 1.** Specific heat divided by the temperature vs.  $T^2$  for  $\text{Ce}_3\text{Bi}_4\text{Pt}_3$  single crystal in magnetic fields between zero and 60 T. Dashed lines are guides to the eye.

During magnetic field sweep, the temperature of the heat capacity stage does not stay constant even in the total absence of eddy current heating, due to the magnetocaloric effect. The heat capacity stage is thermally isolated from the bath, and remains in adiabatic condition during the magnetic field pulse. Such a system warms as the field is ramped up, and cools during the ramp down portion of the magnetic field pulse *reversibly*. However, magnetization in general can also increase with temperature. The sample then would cool during the ramp up, and warm *reversibly* during the ramp down of the magnetic field. We are using this effect to analyze the specific heat data collected  $\text{UBe}_{13}$ , a possible electric quadrupolar Kondo system.

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## Specific Heat of $\text{Ce}_{0.8}\text{La}_{0.2}\text{Al}_3$ in Magnetic Fields

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$\text{CeAl}_3$  was the first heavy fermion compound identified, and it was long considered a canonical example of a non-magnetic Kondo lattice system. However, this description has been challenged by the discovery of anomalies in muon spin rotation, nuclear magnetic resonance, and specific heat data below 2 K. Partial replacement of Ce by La enhances a peak in the specific heat, and moves it from 0.4 K in  $\text{CeAl}_3$ , to 2.3 K in  $\text{Ce}_{0.8}\text{La}_{0.2}\text{Al}_3$ . The anomalous features of pure and La-doped  $\text{CeAl}_3$  have variously been attributed to short-range magnetic order, long-range ordering of very small moments, and most recently the anisotropic Kondo effect.<sup>1</sup>

We have measured the specific heat of  $\text{Ce}_{0.8}\text{La}_{0.2}\text{Al}_3$  in magnetic fields up to 14 T. The field weakly depresses the temperature of the specific heat peak, but strongly reduces its height. These trends are qualitatively at odds with numerical results calculated for the anisotropic Kondo model, thereby casting considerable doubt on this theoretical description.<sup>2</sup> At the same time, a sublinear field dependence of the peak temperature contrasts with the superlinear dependences found in other heavy fermion systems that exhibit small-moment magnetism. This leaves open short-range magnetic order as the most plausible explanation of the puzzling behaviors of  $\text{Ce}_{0.8}\text{La}_{0.2}\text{Al}_3$  and its parent compound  $\text{CeAl}_3$ . Muon spin rotation measurements are planned to test this possibility.

**Acknowledgements:** This work was supported in part by DOE grant DE-FG02-99ER45748.

<sup>1</sup> Goremychkin, E.A., *et al.*, Phys. Rev. Lett., **84**, 2211-2214 (2001).

<sup>2</sup> Pietri, R., *et al.*, Phys. Rev. Lett., **86**, 1090-1093 (2001).

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## Preliminary dHvA Measurements of $\text{PuX}_3$ , X=Sn, Ga, and In

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Despite its rather long history and technological importance, the solid state properties of Pu and its compounds remain mysterious. Elemental Pu has the largest electronic coefficient of specific heat of any element in the periodic table, undergoes six structural phase transitions on cooling from its anomalously low melting temperature, and expands on cooling in two of its structural phases. None of these effects are well understood, but the general consensus is that they are controlled by the 5f electrons and their hybridization with Pu's d electrons. Very recent photoemission and band structure calculations support this belief.

At LANL, we have succeeded in growing single crystals of  $\text{PuX}_3$  (X=Sn, In, and Ga) for the first time and have attempted dHvA measurements using the pulsed fields available at NHMFL-LANL to address these issues and to study the Fermi surfaces of Pu and its compounds. Because of the rapid rate of crystal damage due to self radiation, these measurements are only feasible when both the synthesis and the dHvA measurements can be performed at the same facility, as is possible at LANL. Although we were able to demonstrate all the technical steps in preparing the crystals, encapsulating samples, transferring them to NHMFL, mounting on an appropriate probe, and performing pulsed-field magnetization measurements at cryogenic temperatures, our first attempts to observe quantum oscillations in  $\text{PuX}_3$  failed due to the self damage effects alluded to above. The samples were transferred to NHMFL-LANL just prior to the Cerro Grande fire in May, and the closure of the laboratory for an extended period resulted in crystals with inadequate quality. Further measurements on newly prepared samples are now in progress.

## Electrical Resistivity of $\text{UBe}_{13}$ in High Magnetic Fields: Aging Effects

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We have previously reported efforts to characterize the ground state of the heavy fermion superconductor  $\text{UBe}_{13}$  with electrical resistivity measurements in high magnetic fields.<sup>1</sup> These measurements focused on the region above 12 T using a resistive magnet. During the past year we sought to complement this study with measurements made in fields below 18 T using a superconducting magnet. We used the same high quality single crystal sample and the same sample leads (the contact resistance per lead was less than, or about 1  $\Omega$ ). These new measurements were made 2.0 years after the original measurements.

We sought to determine the field dependence of the boundary between the low temperature Fermi-liquid state and the higher temperature non-Fermi-liquid state. Our earlier measurements were confirmed in that the boundary showed very little field dependence, but the boundary occurred at a temperature significantly lower than that of our earlier study: from about 0.8 K to 0.55 K at 12 T. Subsequent measurements showed that during this same two-year interval the superconducting critical temperature increased from 0.86 to 0.91 K, and that the upper critical field increased from 9.0 to 9.8 T (below 50 mK). All of these changes were larger than the experimental uncertainty of the measurements.

It has been recognized that alpha decay of the uranium in  $\text{UBe}_{13}$  and  $\text{UPt}_3$  can give rise to sample dependent properties,<sup>2</sup> but we are unaware of any specific studies of aging effects in these materials. It is interesting to speculate that the increased disorder created by the alpha decay may be responsible for strengthening the non-Fermi-liquid state and thus reducing the crossover temperature. Further work is required to clarify these issues.

<sup>1</sup> Schmiedeshoff, G.M., *et al.*, NHMFL Research Reports 1999.

<sup>2</sup> Smith, J.L., *Philosophical Magazine B*, **65**, 1371 (1992).

## Ultrasonic Spectroscopy in Pulsed Magnetic Fields

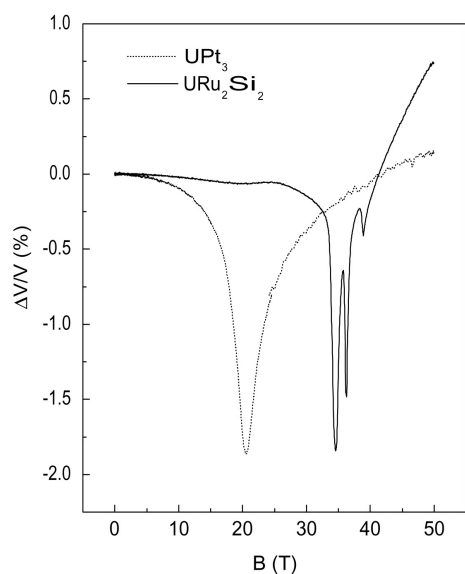
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A fast data acquisition ultrasonic spectrometer for experimental measurements in pulsed magnets has been built at UWM and tested at the NHMFL/Los Alamos. The experimental set up allows us to measure ultrasound velocity and attenuation in the range 10 to 1000 MHz. The spectrometer is intended for investigating the properties of strongly correlated materials at high magnetic fields.

A rf signal from a generator is pulsed and sent through the sample, where it is converted to an acoustic pulse via a piezoelectric transducer bonded to one side of the sample. An echo train is generated and detected at the other end by a second transducer. This received rf signal is amplified and mixed with a local oscillator, the output of which is further amplified and applied to a phase sensitive detector with in-phase and quadrature outputs. The reference signal for the phase sensitive detector is obtained by independently mixing the second channel from the synchronizer with the local oscillator. The amplitude and phase are then computed, yielding immediate changes in the attenuation and velocity.

For continuous magnetic fields, where the magnetic field can be varied slowly, signal averaging with a box-car can improve the sensitivity of the

measurements. For the pulsed magnets, however, data for each individual pulse is stored directly into a computer or digital oscilloscope. A train of a large number of pulses is sent in rapidly during the duration of the magnet pulse, and all the information stored. During the time the capacitor banks recharge and the magnet cools down, the data is transferred from the memory banks to the hard disk in the computer. A high power rf amplifier is needed to ensure that the average pulse height does not fall off during the train of pulses. To reduce eddy current heating, the sample holder is made of G-10, and a minimal amount of metal is used in the vicinity of the sample. The signal to noise is thus not comparable to the case of the continuous magnetic fields.



**Figure 1.** Ultrasonic velocity at the metamagnetic transition in  $\text{UPt}_3$  and  $\text{URu}_2\text{Si}_2$  in a pulsed magnetic field.

Measurements have been performed on two heavy fermion crystals,  $\text{UPt}_3$  and  $\text{URu}_2\text{Si}_2$ , in the short pulsed (25 ms) 50 T magnet. The field is measured by winding a small coil (about ten turns) on the sample holder, and integrating the induced emf. Fig. 1 shows a representative graph of the relative velocity change as a function of magnetic field. In  $\text{UPt}_3$ , the magnetic field is applied along the b-axis; and in  $\text{URu}_2\text{Si}_2$ , the field is applied along the c-axis. Measurements in both crystals were done in the vicinity of 2 K in a pumped He cryostat. In  $\text{UPt}_3$ , the metamagnetic dip in the velocity is seen at 20 T. The ultrasonic pulses were at a frequency of 25 kHz (40  $\mu\text{s}$  between

pulses). There is enough resolution to follow through the dip in the velocity. Measurements on a single crystal of  $\text{URu}_2\text{Si}_2$  were also done in the pulsed magnetic field. The field was applied along the c-axis. Metamagnetic behavior, similar to that in  $\text{UPt}_3$ , is also seen. The data show a 3-fold splitting.

**Acknowledgements:** This work was supported by NSF DMR-9704020 and DMR-9971123.

## Non-Fermi Liquid Behavior at a Metamagnetic Transition?/Specific Heat and Magnetization as a Function of Field; Specific Heat in Field of $\text{CeTIn}_5$

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Work on C(H) and M(H) (the latter using the Vibrating Sample Magnetometer at NHMFL, Tallahassee) in fields up to 20 T and 37 T respectively was performed on single crystals of  $\text{UPd}_2\text{Al}_3$  ( $H_{\text{metamag}}=18$  T) and  $\text{URu}_2\text{Si}_2$  ( $H_{\text{metamag}}\approx 36$  T) to look for non-Fermi liquid (nFl) behavior at the metamagnetic transition as found<sup>1</sup> last year by our group at  $H_{\text{metamag}}=20.5$  T in  $\text{UPt}_3$ . The results on  $\text{UPd}_2\text{Al}_3$  indicate that no temperature-dependent long range interactions are present at  $H_{\text{metamag}}$ , i. e. no nFl behavior is observed in the specific heat. An interesting behavior of the antiferromagnetism with field up to 18.5 T is observed, followed by a change in behavior at higher fields.<sup>2</sup> Initial magnetization results on  $\text{URu}_2\text{Si}_2$  up to 37 T, however, on  $\text{URu}_2\text{Si}_2$  indicate the possibility of nFl behavior at  $H_{\text{metamag}}$ .

Specific heat as a function of field up to 32 T was measured on single crystals of  $\text{CeTIn}_5$ , T=Co, Rh, and Ir. For T=Ir, the upturn in C/T in zero field for T<5 K is suppressed such that by 23 T,  $\text{CeIrIn}_5$  appears to be a normal metal with, however, an enhanced  $\gamma$  of  $\sim 400$  mJ/mol-K<sup>2</sup>. Similar results for  $\text{CeCoIn}_5$  were found. In the antiferromagnetic  $\text{CeRhIn}_5$ ,  $T_N$  decreased from 3.7 K in zero field to 2.1 K in 32 T and the upturn in C/T as temperature is decreased toward  $T_N$  is reduced by  $\sim 50\%$  by 32 T.



**Acknowledgements:** Work at the University of Florida is supported by DOE contract #DE-FG05-86ER45268.

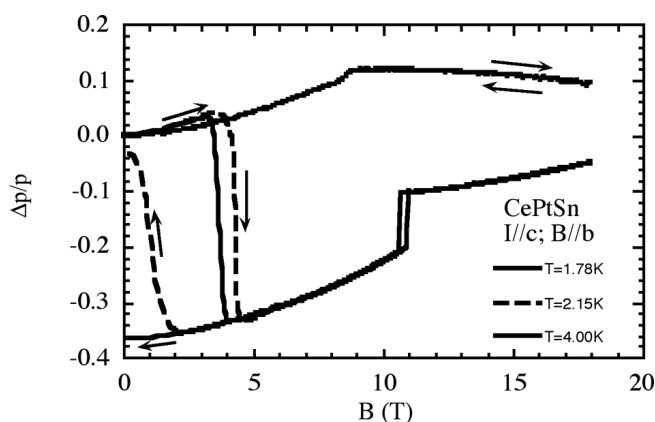
<sup>1</sup> Kim, J.S., *et al.*, Solid State Comm. **114**, 413 (2000).

<sup>2</sup> Kim, J.S., *et al.*, to be published.

## Irreversible Magnetoresistance in Single-Crystalline CePtSn

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The Kondo-lattice compound CePtSn displays remarkable electronic and magnetic properties at low temperatures, which originate in the competition between the Kondo and Ruderman-Kittel-Kasuya-Yosida (RKKY) ground states. This compound crystallizes in the orthorhombic  $\epsilon$ -TiNiSi-type structure. The CePtSn compound is particularly interesting because of the complexity of its magnetic phase diagram,<sup>1</sup> and the observation of spin-slip in the magnetically ordered phases,<sup>2</sup> reminiscent to Ho or PrGa<sub>2</sub>.



**Figure 1.** Transverse magnetoresistance vs. temperature in CePtSn.

In the spin-slip phase, the ionic moments are confined to the easy axis direction, yielding a local magnetic structure that is simple and commensurate. However, the occurrence of periodic spin slips renders the

structure incommensurate on a large scale. In this experiment, we performed measurements of transverse magnetoresistance MR with  $I//c$ -axis, and  $B//b$ -axis. The data of Fig. 1 show that the MR at 1.78 K drops sharply at about 3 T, and that this transition is irreversible with field. At a slightly high temperature,  $T=2.15$  K, the drop in MR takes place at a slightly higher field of 4 T. It is still irreversible, but it starts to increase again near 2 T upon lowering  $B$ . At  $T=4$  K, there are no signs of this transition. It is tempting to ascribe the drop in MR at  $B=3$  to 4 T for  $T<2.15$  K to the magnetic field annealing of the spin-slip phase. Further experiments are in order.

<sup>1</sup> Takabatake, T., *et al.*, Physica B, **183**, 108 (1993).

<sup>2</sup> Kadowaki, H., J. Phys. Soc. Jpn., **67**, 3261 (1998).

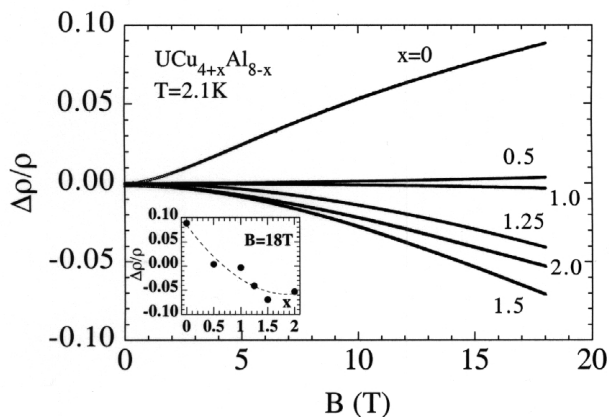
## Magnetoresistance in $\text{UCu}_{4+x}\text{Al}_{8-x}$ Compounds

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The  $\text{UCu}_{4+x}\text{Al}_{8-x}$  ( $0.1 \leq x \leq 1.95$ ) compounds crystallize in the tetragonal  $\text{ThMn}_{12}$ -type structure.<sup>1</sup> The Cu-poor compounds within this  $x$ -range are AF with  $T_N$  near 35 K. However, the substitution of Cu for Al is accompanied by (1) a rapid suppression of AF in the  $0.1 < x < 1.15$  range, and (2) an enormous increase in  $\gamma$  from about 100 mJ/mole-K<sup>2</sup> ( $x \approx 0.25$ ) to 750 mJ/mole-K<sup>2</sup> ( $x = 1.75$ ).<sup>2</sup> Neutron diffraction results suggest that the substitution of Cu for Al favors one of two inequivalent Al sites.<sup>3</sup> Gschneidner *et al.* pointed out that false indications of heavy fermion HF behavior can occur as a result of non-magnetic atomic disorder.<sup>4</sup>

In order to probe further the effect of the substitution of Cu for Al in  $\text{UCu}_{4+x}\text{Al}_{8-x}$ , we performed a series of measurements of  $\chi$  vs.  $T$  and magnetoresistance

MR in B up to 18 T. The behavior of the transverse MR vs. B at T=2.1 K for the six samples of this study is displayed in Fig. 1. The MR of the multiphase compound  $\text{UCu}_4\text{Al}_8$  is positive. However, as more Cu is substituted for Al forming single-phase materials, the MR becomes negative, and its magnitude grows from near zero, for  $x=0.5$ , to a maximum near  $x=1.5$ , diminishing again for multiphase  $x=2.0$ . The MR for the  $0.5 \leq x \leq 2.0$  compositions follows closely a  $B^2$  dependence.



**Figure 1.** Transverse magnetoresistance  $\Delta\rho/\rho$  vs. B for  $\text{UCu}_{4+x}\text{Al}_{8-x}$  compounds at 2.1 K. The inset shows  $\Delta\rho/\rho$  vs. x at B=18 T. The line is a guide to the eye.

The behavior of  $\Delta\rho/\rho$  vs. B at higher T is qualitatively similar, albeit reduced in magnitude. The MR in strongly correlated systems at low T can usually be described by  $\Delta\rho(B,T) = \Delta\rho_c(B,T) + \Delta\rho_{sf}(B,T)$ , where the first term is a positive contribution due to the cyclotron motion of the charge carriers, and the second term is a negative contribution due to the suppression of spin fluctuations with B. The  $\Delta\rho/\rho$  vs. B data of Fig. 1 suggest that the cyclotron contribution from the impurities is larger than the spin-fluctuation term in  $\text{UCu}_4\text{Al}_8$ . Upon the substitution of more Cu for Al, the compounds become single-phase, and the MR becomes progressively more negative, suggesting that spin fluctuation scattering becomes more relevant when  $T_N$  is reduced or suppressed. The negative value of  $\Delta\rho/\rho$ , the quadratic behavior with B, and the drop in magnitude with T are consistent with the spin fluctuation theory developed by Ueda.<sup>5</sup> The important question of whether these materials

are real or false HF remains, and more studies are in order.

- <sup>1</sup> Geibel, C., *et al.*, Physica B, **163**, 194 (1990).
- <sup>2</sup> Nishioka, T., *et al.*, Czech. J. Phys., **46**, 2065 (1996).
- <sup>3</sup> Krimmel, A., *et al.*, J. Magn. Magn. Mat., **104-107**, 25 (1992).
- <sup>4</sup> Gschneidner, Jr., K.A., *et al.*, Physica B, **163**, 507 (1990).
- <sup>5</sup> K. Ueda, Solid State Commun. **19**, 965 (1976).

## Investigation of Low-Temperature Resistivity of the Non-Fermi Liquid Compound $\text{Sc}_{1-x}\text{U}_x\text{Pd}_3$

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Recent measurements in our laboratory indicate that the  $\text{Sc}_{1-x}\text{U}_x\text{Pd}_3$  system exhibits non-Fermi liquid behavior for uranium concentrations near  $x=0.3$ . The temperature dependence of the electrical resistivity of samples with x near 0.3 at very low temperatures follows a power law  $\rho \sim T^n$  with  $n=0.5$  in good agreement with the predictions of the two-channel, spin-1/2, electric quadrupolar Kondo effect. It is predicted that the electrical resistivity of these compounds should cross over to Fermi-liquid behavior ( $\rho \sim T^n$  with  $n=2$ ) at high magnetic fields. We have measured the temperature dependence of the electrical resistivity in magnetic fields at NHMFL/LANL and have found that a cross over to Fermi-liquid behavior is not present in fields up to 18 T.